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(54) Title: LUBRICATION OIL COMPOSITION

(57) Abstract

A lubricating oil composition comprising (A) a base oil containing 3 % by weight or less of aromatics, 20 % by weight or more of monocyclic naphthenes, 50 ppm by weight or less of N and 50 ppm by weight or less of S, having a viscosity (100 °C) of 2 to 50 mm^2/s, (B) alkyldiphenylamines and/or phenyl-α-naphthylamines in an amount of 0.05 to 2 % by weight of the total weight of the composition, and (C) C_{8-23} MoDTC and/or C_{8-18} MoDTP and/or C_{8-18} MoDTX in such an amount that the amount of Mo is 50 to 2000 ppm by weight of the total weight of the composition.

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LUBRICATION OIL COMPOSITION

BACKGROUND OF THE INVENTION

Field Of The Invention

The present invention relates to a novel lubricating oil composition, and more specifically to a lubricating oil composition having high heat resistance, high oxidation stability and excellent lubricating properties, useful as a lubricating oil for internal-combustion engines, automatic transmission gearboxes, dampers, power steerings and the like, particularly useful as a lubricating oil for internal combustion engines.

Description Of The Related Art

Heretofore, lubricating oils have been used for internal combustion engines, and for driving units and gears such as automatic transmission gearboxes, dampers and power steerings in order to smoothly operate them. In particular, lubricating oils for internal-combustion engines (engine oils) not only lubricate various sliding portions such as a piston ring, a cylinder liner, bearings for a crank shaft and a connecting rod, and a valve-operating mechanism including a cam and a valve lifter, but also cool the inside of the engines, clean and disperse those products which are produced by combustion, and prevent the rusting and corrosion of the engines.

Thus, the lubricating oils for internal-combustion engines have been required to have a great variety of properties. Moreover, due to the recent trend toward high-performance, high-output internal-combustion engines and more severe operating conditions, the lubricating oils are required to have higher quality. In order to meet this requirement, various additives such as an antiwear agent, a metallic detergent, a nonash dispersant and an antioxidant are incorporated into the lubricating oils for internal-combustion engines.

It is particularly important as the essential function of the lubricating oils for internal-combustion engines that the lubricating oils can ensure the smooth operation of the engines under every conditions to prevent the wear and seizure of the engines. The parts of the engines to be lubricated are, in most cases, under the fluid

lubrication condition. However, a valve-operating system, and the top and bottom dead centers of a piston tend to be under the boundary lubrication condition. Antiwear properties under the boundary lubrication condition are generally imparted by the addition of zinc dithiophosphate (ZnDTP) or zinc dithiocarbamate (ZnDTC).

Energy loss at the frictional parts of internal-combustion engines which are lubricated by lubricating oils is great. For this reason, a lubricating oil to which various additives including a friction modifier (FM) are added has been used in order to reduce the friction loss and to decrease the fuel cost (e.g., Japanese Laid-Open Patent Publication No. 23595/1991). Lubricating oils for automotive internal-combustion engines are used at various temperatures, with various numbers of revolutions and under various loads. Therefore, in order to further improve the rate of fuel consumption, it is necessary that the lubricating oils be excellent in friction properties under a wide range of conditions under which they are used.

Besides the above-described properties, high heat resistance, high oxidation stability and moderate viscosity characteristics can be mentioned as the properties required for the lubricating oils for internal-combustion engines.

SUMMARY OF THE INVENTION

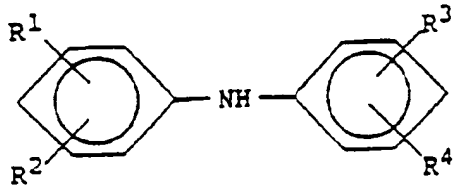
The present invention has been accomplished under these circumstances. An object of the present invention is therefore to provide a lubricating oil composition having excellent lubricating properties, high heat resistance, high oxidation stability and moderate viscosity characteristics, particularly useful as a lubricating oil for internal-combustion engines.

We have made earnest studies in order to develop a lubricating oil composition having the above-described advantageous properties, and, as a result, found that the object can be attained by a composition which is obtainable by adding a predetermined amount of a specific amine antioxidant, and a predetermined amount of oxymolybdenum sulfide dithiocarbamates (MoDTC), oxymolybdenum sulfide organo-phosphorodithioates (MoDTP) or oxymolybdenum sulfide dithioxanthogenates (MoDTX) to a lubricating base oil containing a small amount of aromatics, having specific characteristics. The present invention has been accomplished on the basis of the above finding.

Namely, the present invention relates to a lubricating oil composition comprising:

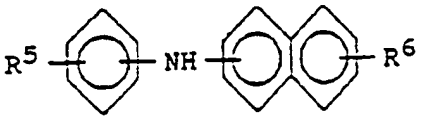
(A) a lubricating base oil containing 3% by weight or less of aromatics, 20% by weight or more of monocyclic naphthenes, 50 ppm by weight or less of sulfur and 50 ppm by weight or less of nitrogen, having a viscosity of 2 to 50 mm^2/s at 100°C;

(B) at least one compound selected from diarylamines represented by the general formula [1]:



[1]

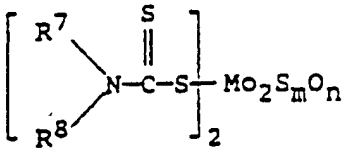
wherein R^1, R^2, R^3 and R^4, which may be the same or different, each represent hydrogen atom or a hydrocarbon group having 3 to 18 carbon atoms, provided that at least one of them is the hydrocarbon group, and by the general formula [2]:



[2]

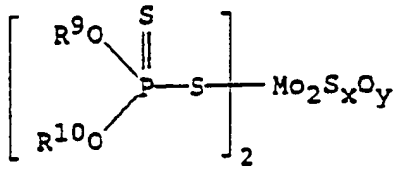
wherein R^5 and R^6 are hydrogen atom or a hydrocarbon group having 1 to 18 carbon atoms, in an amount of 0.05 to 3% by weight of the total weight of the composition; and

(C) at least one compound selected from oxymolybdenum sulfide dithiocarbamates represented by the general formula [3]:



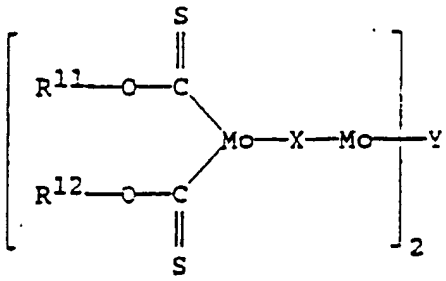
[3]

wherein R^7 and R^8, which may be the same or different, each represent a hydrocarbon group having 5 to 23 carbon atoms, and m and n are a positive integer, provided that the total number of m and n is 4, oxymolybdenum sulfide organophosphorodithioates represented by the general formula [4]:



[4]

wherein R^9 and R^{10}, which may be the same or different, each represent a hydrocarbon group having 1 to 18 carbon atoms, and x and y are a positive integer, provided that the total number of x and y is 4, and oxymolybdenum sulfide dithioxanthogenates represented by the general formula [5]:



[5]

wherein R^{11} and R^{12}, which may be the same or different, each represent a hydrocarbon group having 1 to 30 carbon atoms, and X and Y, which may be the same or different, each represent oxygen or sulfur atom,

in such an amount that the amount of molybdenum is 50 to 2000 ppm by weight of the total weight of the composition.

BRIEF DESCRIPTION OF THE DRAWING

Figure 1 is a diagrammatic view of an apparatus used in the LFW-1 friction test.

DESCRIPTION OF THE PREFERRED EMBODIMENTS

In a preferred embodiment of the present invention, the lubricating base oil is a hydrogenated oil containing 3% by weight or less of aromatics, 20% by weight

or more of monocyclic naphthenes, and 97% by weight or more of saturated compounds.

In another preferred embodiment, the lubricating base oils is a hydrogenated oil, the diarylamines are alkyldiphenylamines containing at least one alkyl group having 3 to 18 carbon atoms or phenyl-α-naphthylamines containing an alkyl group having 3 to 18 carbon atoms, and the third component is an oxymolybdenum.

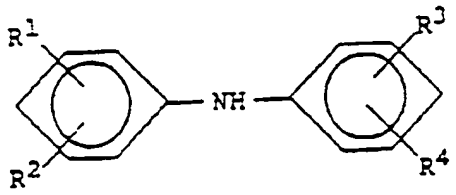
In yet another preferred embodiment, the oxymolybdenum sulfide organophosphorodithioates contain an alkyl group having 8 to 18 carbon atoms and/or the oxymolybdenum sulfide dithioxanthogenates contain an alkyl group having 8 to 18 carbon atoms.

Still another preferred embodiment relates to a method for reducing fuel consumption by the use of the lubricating oil composition according to the invention in an engine.

In the lubricating oil composition of the present invention, an oil containing 3% by weight or less of aromatics, 20% by weight or more of monocyclicnaphthenes, 50 ppm by weight or less of sulfur and 50 ppm by weight or less of nitrogen, having a viscosity of 2 to 50 mm^2/s at 100°C is used as the lubricating base oil, the component (A). The preferable amount of the monocyclic naphthenes is in the range of 25 to 40% by weight. When the amount of the aromatics exceeds 3% by weight, the resulting lubricating oil composition undergoes deterioration in heat resistance, oxidation stability and lubricating properties. In the case where the amount of the monocyclic naphthenes is less than 20% by weight, the resulting composition cannot have sufficiently high adaptability to sealing rubber. Further, when the lubricating base oil has a viscosity of lower than 2 mm^2/s, the resulting composition is poor in the oil-film-forming properties, and has a shortcoming in that it undergoes a great evaporation loss. A base oil having a viscosity of higher than 50 mm^2/s is also unfavorable because the power loss of the resulting composition caused by viscosity resistance is too great. Furthermore, when either sulfur or nitrogen content exceeds 50 ppm by weight, the oxidation stability and lubricating properties of the resulting composition become poor.

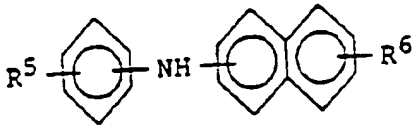
Either mineral or synthetic oil can be used as the lubricating base oil as long as it has the aforementioned properties. Specific examples of the base oil include raffinates which can be obtained by subjecting starting materials for lubricating oils derived from naphthene base or paraffin base crude oil by evaporation under normal or reduced pressure to solvent refining, using an aromatic extraction solvent such as phenol, furfural or N-methylpyrrolidone, and hydrogenated oils which can be obtained by subjecting starting materials for lubricating oils to hydrogenation treatment including hydrocracking reaction. In either production process, such processes as dewaxing, hydrorefining and clay treatment processes may be optionally adopted in accordance with the conventional manner. Particularly preferably base oils are hydrocracked oils and wax-isomerized oils.

In the composition of the present invention, at least one compound selected from diarylamines represented by the general formula [1]:



[1]

or by the general formula [2]:



[2]

is used as the amine oxidant, the component (B).

In the above general formula [1], R^1, R^2, R^3 and R^4 each represent hydrogen atom or a hydrocarbon group having 1 to 18 carbon atoms. Further, although R^1, R^2, R^3 and R^4 may be the same or different from one another, it is necessary that at least one of them be an alkyl group having 3 to 18 carbon atoms. The alkyl group having 3 to 18 carbon atoms may be any of linear, branched and cyclic ones. Examples of such an alkyl group include propyl, butyl, amyl, hexyl, heptyl, octyl, nonyl, decyl, undecyl, dodecyl, tridecyl, tetradecyl, pentadecyl, hexadecyl,

heptadecyl and octadecyl groups of all types, and cyclohexyl, cyclooctyl and cyclododecyl groups.

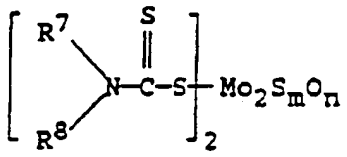
In the above general formula [2], R^5 and R^6 are hydrogen atom or a hydrocarbon group having 1 to 18 carbon atoms. A preferable hydrocarbon group is an alkyl group having 3 to 18 carbon atoms, which maybe any of linear, branched and cyclic ones. Examples of such an alkyl group include the same groups as those enumerated in the explanation of R^1, R^2, R^3 and R^4 in the above general formula [1]. Specifically, the following compounds can be mentioned as the diarylamines: p,p'dibutyl-diphenylamine, p,p'-dipentyldiphenylamine, p,p'-dihexyldiphenylamine, p,p'-diheptyldiphenylamine, p,p'-dioctyldiphenylamine, p,p'-dinonyldiphenylamine, monooctyldiphenyl-amine, monononyldiphenylamine, tetrabutyldiphenylamine, tetrahexyldiphenylamine, tetraoctyldiphenylamine, tetranonyldiphenylamine, a mixture of alkyldiphenylamines having 4 to 9 carbon atoms, phenyl-α-naphthylamine, phenyl-β-naphthylamine, butylphenyl-α-naphthylamine, butylphenyl-β-naphthylamine, pentylphenyl-α-naphthylamine, pentylphenyl-β-naphthylamine, hexylphenyl-α-naphthylamine, hexylphenyl-β-naphthylamine, heptylphenyl-α-naphthylamine, heptylphenyl-β-naphthylamine, octylphenyl-α-naphthylamine, octylphenyl-β-naphthylamine, nonylphenyl-α-naphthylamine and nonylphenyl-β-naphthylamine. Particularly preferable diarylamines are p,p'-dioctyldiphenylamine, phenyl-α-naphthylamine and alkylphenyl-α-naphthylamines.

In the composition of the present invention, one or two or more of the alkyldiphenylamines represented by the above general formula [1], or one or two of more of the phenyl-α-naphthylamines represented by the above general formula [2] may be used as the amine oxidant, the component (B). Moreover, one or more of the alkyldiphenylamines represented by the general formula [1], and one or more of the phenyl-α-naphthylamines represented by the general formula [2] may also be used in combination as the amine antioxidant.

In the present invention, it is necessary to incorporate the amine antioxidant, the component (B), into the composition in an amount of 0.05 to 3% by weight, preferably 0.2 to 2% by weight of the total weight of the composition. When the amount of the amine antioxidant is less than 0.05% by weight, the resulting composition cannot have sufficiently high oxidation stability. On the other hand, when

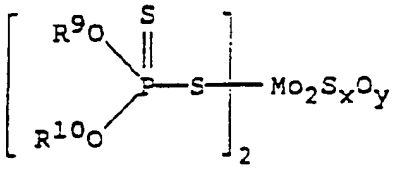
the amount is in excess of 3% by weight, the effects of the antioxidant expected from such an amount cannot be obtained.

In the composition of the present invention, at least one compound selected from oxymolybdenum sulfide dithiocarbamates (MoDTC) represented by the general formula [3]:



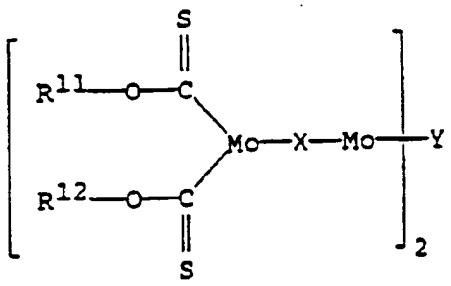
[3]

oxymolybdenum sulfide organophosphorodithioates (MoDTP) represented by the general formula [4]:



[4]

and oxymolybdenum sulfide dithioxanthogenates (MoDTX) represented by the general formula [5]:



[5]

is used as the friction modifier, the component (C).

In the above general formula [3], R^7 and R^8 each represent a hydrocarbon group having 5 to 23 carbon atoms, and they may be the same or different from each other. Examples of the hydrocarbon group having 5 to 23 carbon atoms include a linear or branched alkyl or alkenyl group having 5 to 23 carbon atoms, and a cycloalkyl, aryl, alkylaryl or arylalkyl group having 6 to 23 carbon atoms. Preferable hydrocarbon groups are those having 8 to 23 carbon atoms. Specific examples of such hydrocarbon groups include 2-ethylhexyl, n-octyl, nonyl, decyl, lauryl, tridecyl,

palmityl, stearyl, oleyl, eicosyl, butylphenyl and nonylphenyl groups. Further, m and n are a positive integer, provided that the total number of m and n is 4.

In the above general formula [4], R^9 and R^{10} each represent a hydrocarbon group having 1 to 18 carbon atoms, and they may be the same or different from each other. Preferable hydrocarbon groups are those having 3 to 18 carbon atoms. Examples of the hydrocarbon groups having 3 to 18 carbon atoms include a linear or branched alkyl or alkenyl group having 3 to 18 carbon atoms, a cycloalkyl group having 6 to 18 carbon atoms, an aryl group having 6 to 18 carbon atoms, and an alkylaryl or arylalkyl group having 7 to 18 carbon atoms. Specific examples of such groups include isopropyl, n-propyl, n-butyl, isobutyl, sec-butyl, amyl, hexyl, cyclohexyl, 2-ethylhexyl, n-octyl, nonyl, decyl, lauryl, tridecyl, palmityl, stearyl, oleyl, butylpheyl and nonylphenyl groups. Further, x and y are a positive integer, provided that the total number of x and y is 4.

In the above general formula [5], R^{11} and R^{12} each represent a hydrocarbon group having 1 to 30 carbon atoms, and they may be the same or different from each other. Preferable hydrocarbon groups are those having 3 to 20 carbon atoms. Examples of such hydrocarbon groups include a linear or branched alkyl or alkenyl group having 5 to 20 carbon atoms, a cycloalkyl group having 6 to 20 carbon atoms, and an aryl, alkylaryl or arylalkyl group having 6 to 20 carbon atoms. Specific examples of such groups include isopropyl, n-propyl, isobutyl, n-butyl, sec-butyl, amyl, hexyl, cyclohexyl, 2-ethylhexyl, n-octyl, nonyl, decyl, lauryl, tridecyl, palmityl, stearyl, oleyl, butylpheyl and nonylphenyl groups. Further, X and Y are oxygen or sulfur atom, and may be the same or different from each other.

In the composition of the present invention, the MoDTC represented by the above general formula [3] may be used either singly or in combination of two or more. The MoDTP represented by the general formula [4] may also be used either singly or in combination of two or more. Further, the MoDTX represented by the general formula [5] may also be used either singly or in combination of two or more.

In the composition of the present invention, it is necessary to incorporate the friction modifier, the component (C), into the composition in such an amount that the amount of molybdenum will be 50 to 2000 ppm by weight, preferably 100 to 1000 ppm by weight of the total weight of the composition. When the amount

of molybdenum is less than 50 ppm by weight, lubricating properties cannot be sufficiently obtained. On the other hand, when the amount of molybdenum is in excess of 2000 ppm by weight, lubricating properties expected from such an amount cannot be obtained.

Those additives which have been usually incorporated into the conventional lubricating oils, such as a metallic detergent, a nonash detergent-dispersant, an antiwear agent, a viscosity index improver, a pour point depressant, a rust preventive, a corrosion inhibitor, an anti-foaming agent and other antioxidants, can be added, if necessary, to the lubricating oil composition of the present invention within such a limit that the object of the present invention can be fully attained.

Examples of the metallic detergent include calcium sulfonate, magnesium sulfonate, barium sulfonate, calcium phenate, barium phenate, calcium salicylate and magnesium salicylate. In general, the metallic detergent is incorporated into the composition in an amount of 0.1 to 5% by weight. Examples of the nonash detergent-dispersant include those of succinimide type, succinamide type, benzylamine or its boron derivative type and ester type. In general, such a detergent is incorporated into the composition in an amount of 0.5 to 7% by weight.

Examples of the antiwear agent include metallic (Zn, Pb, Sb, Mo, etc.) salts of thiophosphoric acid, metallic (Zn, etc.) salts of thiocarbamic acid, sulfur compounds, phosphoric esters and phosphorous esters. In general, this agent is incorporated into the composition in an amount of 0.05 to 5.0% by weight.

Examples of the viscosity index improver include those of polymethacrylate type, polyisobutylene type, ethylene-propylene copolymer type and styrene-butadiene hydrogenated copolymer type. In general, such an improver is incorporated into the composition in an amount of 0.5 to 35% by weight.

Examples of the rust preventive include alkenyl succinates and partial esters thereof. Examples of the corrosion inhibitor include benzotriazole and benzoimidazole. Examples of the anti-foaming agent include dimethyl polysiloxane and polyacrylate. These agents may be incorporated into the composition, when necessary.

The present invention will now be explained more specifically by referring to the following Examples. However, the present invention is not limited by these examples in any way.

EXAMPLES

The oxidation-induction time and coefficient of friction of the lubricating oil compositions were obtained in the following respective manners.

(1) Coefficient of Friction (μ)

LFW-1 friction test was carried out by using a LFW-1 tester shown in Figure 1 equipped with a R-type block (made of iron) manufactured by Falex Corporation and a S-10 test ring (made of iron) manufactured by Falex Corporation under the following conditions: the number of revolutions was 270 rpm, the load was 30 kgf, the temperature of the oil was 120°C, and the time was 10 minutes. In Figure 1, reference numeral 1 indicates the S-10 test ring, reference numeral 2 indicates the R-type block and reference numeral 3 indicates a distortion meter. Load is applied to the R-type block, and the resistance caused by the rotation of the ring is measured by the distortion meter. The coefficient of friction is calculated from the resistance measured. It is noted that approximately half of the ring is immersed in the oil to be tested.

(2) Oxidation-Induction Time (Minutes)

The oxidation-induction time was determined by means of differential thermal analysis which was conducted under oxygen atmosphere, by applying a load of 20 kgf/cm^2 and heating the sample to 200°C.

Examples 1 to 8 and Comparative Examples 1 to 7

By the use of a base oil shown in Table 1, a lubricating oil composition having a formulation shown in Table 2 was prepared. The coefficient of friction (μ) and oxidation-induction time (minutes) of the composition were obtained. The results are shown in Table 2.

TABLE 1

1 BASE OIL	2 (MM^2/S)	3 (WT%)	4 (WT%)	5 (WT. PPM)	6 (WT. PPM)
70N	3.1	1.1	32	1.0	0.3
150N-1	5.5	0.5	30	0.5	0.1
350N	9	1.3	32	0.1	0.1
150N-2	5.6	4.0	18	13.0	1.0
150N-3	5.4	2.0	24	137.0	71.0
150N-4	57	41	19	11.0	89.0

1. Base oil
2. Viscosity at 100°C (mm^2/s)
3. Aromatic content (wt%)
4. Monocyclic naphthene content (wt%)
5. Sulfur content (wt. ppm)
6. Nitrogen content (wt. ppm)

TABLE 2

			Example 1	Example 2	Example 3	Example 4	Example 5	Example 6	Example 7	Example 8
Chemical Composition (Wt%)	Base Oil	70N	balance	-	-	-	-	-	-	-
		150N-1	-	balance	-	balance	balance	balance	balance	balance
		350N	-	-	balance	-	-	-	-	-
		150N-2	-	-	-	-	-	-	-	-
		150N-3	-	-	-	-	-	-	-	-
		150N-4	-	-	-	-	-	-	-	-
	alkyl (C_{4-8})diphenylamine		0.5	0.5	0.5	1.5	-	-	0.5	0.5
	alkylated (C_8)phenyl-α-naphthylamine		-	-	-	-	0.3	1.2	-	-
	4,4'-methylenebis(2,6-di-t-butylphenol)		-	-	-	-	-	-	-	-
	MoDTC (C_{13})		1.0	1.0	1.0	1.0	1.0	1.0	-	-
	MoDTP (C_6)		-	-	-	-	-	-	0.6	-
	MoDTX (C_{18})		-	-	-	-	-	-	-	0.4
Evaluation	coefficient of friction (μ)		0.035	0.039	0.042	0.041	0.040	0.037	0.045	0.041
	oxidation induction period (minutes)		24.3	25.6	29.3	33.6	26.6	36.1	25.9	26.5

TABLE 3

			Comparative Example 1	Comparative Example 2	Comparative Example 3	Comparative Example 4	Comparative Example 5	Comparative Example 6	Comparative Example 7
Chemical Composition (Wt%)	Base Oil	70N	-	-	-	-	-	-	-
		150N-1	-	-	-	-	-	-	-
		350N	-	-	-	-	balance	-	-
		150N-2	balance	-	-	balance	-	-	balance
		150N-3	-	balance	-	-	-	-	-
		150N-4	-	-	balance	-	-	balance	-
	alkyl (C_{4-8})diphenylamine		0.5	0.5	-	0.5	-	-	0.5
	alkylated (C_8)phenyl-α-naphthylamine		-	-	1.2	-	-	-	-
	4,4'-methylenebis(2,6-ti-t-butylphenol)		-	-	-	-	0.5	0.5	-
	MoDTC (C_{13})		1.0	1.0	1.0	-	1.0	1.0	-
	MoDTP (C_6)		-	-	-	0.6	-	-	0.6
	MoDTX (C_{18})		-	-	-	-	-	-	0.4
Evaluation	coefficient of friction (μ)		0.035	0.049	0.051	0.053	0.050	0.056	0.059
	oxidation induction period (minutes)		15.2	14.3	21.8	13.9	21.3	16.4	15.5

All of the lubricating oils of Examples 1 to 8, which are the compositions of the present invention, have a low coefficient of friction and a long oxidation-induction time. In contrast, the lubricating oil of Comparative Example 1 has a low coefficient of friction but has a short oxidation-induction time. The lubricating oils of Comparative Examples 2 to 7 are remarkably inferior to those of Examples in both the coefficient of friction and the oxidation-induction time.

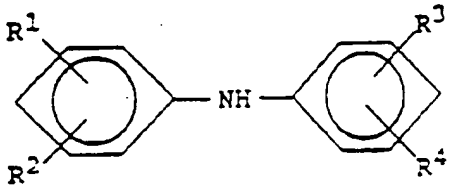
The lubricating oil compositions of the present invention have high heat resistance, high oxidation stability and excellent lubricating properties, and are particularly useful for lubricating oils for internal-combustion engines and the like.

CLAIMS:

1. A lubricating oil composition comprising:

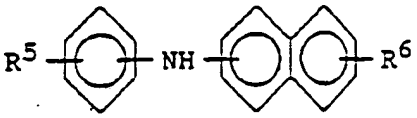
(A) a lubricating base oil containing 3% by weight or less of aromatics, 20% by weight or more of monocyclic naphthenes, 50 ppm by weight or less of sulfur and 50 ppm by weight or less of nitrogen, having a viscosity of 2 to 50 mm^2/s at 100°C;

(B) at least one compound selected from diarylamines represented by the general formula [1]:



[1]

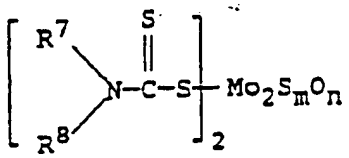
wherein R^1, R^2, R^3 and R^4, which may be the same or different, each represent hydrogen atom or a hydrocarbon group having 1 to 18 carbon atoms, provided that at least one of them is the hydrocarbon group, and by the general formula [2]:



[2]

wherein R^5 and R^6 are hydrogen atom or a hydrocarbon group having 1 to 18 carbon atoms, in an amount of 0.05 to 3% by weight of the total weight of the composition; and

(C) at least one compound selected from oxymolybdenum sulfide dithiocarbamates represented by the general formula [3]:



[3]

INTERNATIONAL SEARCH REPORT

International application No.
PCT/US[redacted]938

A. CLASSIFICATION OF SUBJECT MATTER

IPC(6) :C10M 133/12, 135/14, 135/18, 137/10
US CL : 252/42.7, 50

According to International Patent Classification (IPC) or to both national classification and IPC

B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)

U.S. : 252/42.7, 50

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practicable, search terms used)

APS search terms: diarylamine, lubrica?, dithiocarbamate?, oxymolybdenum

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
Y	US, A, 5,186,852 (ISHIDA ET AL) 16 February 1993, col. 1, lines 52-68, and col. 2, lines 1-44.	1-3 and 6
Y	US, A, 4,832,867 (SEIKI ET AL) 23 May 1989, col. 4, lines 62-68 and col. 5, lines 1-18.	1, 4 and 6
Y	US, A, 4,770,802 (ISHIDA ET AL) 13 September 1988, col. 2, lines 1-43.	1-2 and 6
Y	US, A, 3,840,463 (FROESCHMANN ET AL) 8 OCTOBER 1974, col. 1, lines 46-68.	1, 4 and 6
Y	US, A, 3,356,702 (FARMER ET AL) 5 December 1967, col. 1, lines 44-60.	1
A	US, A, 4,394,279 (deVRIES ET AL) 19 July 1983, See entire document.	1-6

☒ Further documents are listed in the continuation of Box C. ☐ See patent family annex.

* Special categories of cited documents:
"A" document defining the general state of the art which is not considered to be of particular relevance
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"&" document member of the same patent family

Date of the actual completion of the international search	Date of mailing of the international search report
08 MAY 1995	21 JUN 1995
Name and mailing address of the ISA/US Commissioner of Patents and Trademarks Box PCT Washington, D.C. 20231 Facsimile No. (703) 305-3230	Authorized officer ELLEN MCAVOY Telephone No. (703) 308-2510

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